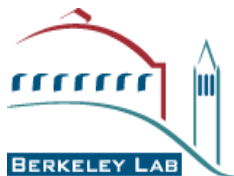




THE GLENN T. SEABORG CENTER
SCIENCE AND EDUCATION FOR HEAVY ELEMENTS AND THE ENVIRONMENT



Actinide Mini-Workshop

Dr. Toshihiko Ohnuki and Dr. Takuo Ozaki
Advanced Basic Research Center, JAERI, Japan

*“Accumulation of Actinides and Lanthanides by
Microorganisms”*

Professor Yngve Albinsson
Chalmers Institute of Technology, Sweden

*“Production of Pu(III), Pu(IV) and Pu(VI) using
Pt/H₂/O₃ in HCl Media”*

Hans Nilsson
Chalmers Institute of Technology, Sweden

“Solubility of Pu(III) hydroxide in Aqueous Solutions”

Monday December 10, 2001
9:15 AM - NOON
Bldg. 70A-Room 3377

Host: Heino Nitsche



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Accumulation of actinides and lanthanides by microorganisms

**Toshihiko OHNUKI and Takuo OZAKI, Advanced Basic Research Center, Japan
Atomic Energy Research Institute**

JAERI has been conducting the biological study to elucidate at the fundamental level the mechanisms of interaction of heavy elements (lanthanides and actinides) with microorganisms. So far, we have investigated the (i) mechanisms of Eu (III) biosorption and (ii) mechanisms of U(VI) bioaccumulation.

In the research for Eu biosorption, we have developed a novel diagram for the characterization of the coordination state of Eu(III) by time-resolved laser-induced fluorescence spectroscopy (TRLFS). Using the diagram, the coordination state of Eu(III) including both the inner- and the outer-sphere can be predicted simply by plotting analytical data for $R_{E/M}$ (the relative intensity of the $^5D_0 \rightarrow ^7F_1$ and $^5D_0 \rightarrow ^7F_2$ transitions) and N_{H_2O} (the number of water molecules in the primary coordination sphere) on the diagram, which is of great advantage in various research fields such as solution chemistry, geological chemistry, and quantum chemistry of lanthanide(III) and actinide(III). In the field of biological chemistry, the diagram would facilitate the characterization of biological molecules containing Eu(III). In the seminar, we would like to present a brief explanation on the diagram and its application to the examination of coordination states of Eu(III) adsorbed onto biopolymers (cellulose, chitin, and chitosan) and bacteria (*Chlorella vulgaris*, *Bacillus subtilis*, *Pseudomonas fluorescens*, *Halomonas* sp., *Halobacterium salinarum*, and *Halobacterium halobium*).

In the research for U bioaccumulation, we have examined the biosorption mechanism of U(VI) by bacteria of *Bacillus subtilis* and *arthrobacter simplex* by batch type isotherm experiments at pHs 4 and 6. Solid phases were analyzed by transmission microscopy using whole mount and thin section samples. We have also examined the biosorption of U(VI) by the mixture of *bacillus subtilis* and kaolinite to elucidate the role of microbe in the migration in terrestrial environment.



Solubility of Plutonium(III) hydroxide in aqueous solutions

Nilsson H., Albinsson Y., Jakobsson A-M., Skarnemark G.

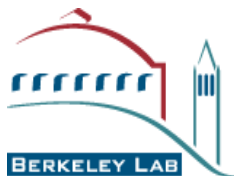
The solubility is an important factor in the migration of radionuclides from a nuclear waste repository. Together with other thermodynamic properties, such as complexation and sorption, it provides an upper limit of the concentrations in the transporting water media. As for plutonium most solubility studies have been performed using compounds with the plutonium present as Pu(IV). PuO₂, hydrated PuO₂ and Pu(OH)₄ are examples of these compounds. However, in the proposed Swedish repository, KBS-3, used nuclear fuel is intended to be disposed of in copper canisters with an interior steel lining. These canisters will be put in bentonite filled tunnels 500 m down in the bedrock. The lack of oxygen in natural waters at this depth, production of H₂ and corrosion of the steel lining are factors that indicate that the redox conditions of the repository will be reducing. Therefore solubility studies of Pu(III) compounds are of interest and they are rare in the literature. In the newly published NEA thermodynamic database of neptunium and plutonium only one reference of Pu(OH)₃ solubility is cited. Our work is intended to further investigate this solubility.

For the solubility experiments we have a nitrogen atmosphere glove box equipped with an ultracentrifuge and cuvette holders connected to an outside-box spectrophotometer via optical fibers. Other box features are a small water-cooled oven, various gas and electrical throughputs, a microscope and a fumehood for organic solvents. Plutonium concentrations were measured using ICP-MS and LSC.

In our preliminary work we used a lead column to reduce a stock solution of Pu to Pu(III). The reduced solution was then put in deoxygenated NaOH to produce a precipitate of Pu(OH)₃. The resulting precipitate, which was creamy brownish-white, was washed and put in solutions of 0.1M NaCl of which were adjusted to different pH. The results show a lower solubility than expected from the K_s reported by Felmy et al. Furthermore speciation and E_H measurements indicate that the plutonium in solution is in the pentavalent or hexavalent state.



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Production of Pu(III), Pu(IV) and Pu(VI) using Pt/H₂/O₃ in HCl media

Y. Albinsson, C. Ekberg, H. Nilsson, A.-M. Jakobsson and G. Skarnemark

To be able to study the behaviour of plutonium in different oxidation states (III, IV, VI) it is essential to be able to produce the different oxidation state as pure as possible. There are several methods in the literature, but most of them include addition of chemicals (oxidation or reduction agents) in order to reduce or oxidize Pu.

In this study only hydrogen gas or ozone has been used together with a platinum catalyst. Thus, chemically very pure plutonium stocks of the different oxidation states can be produced. Absorption spectra (400-900 nm) of the different stocks produced will be shown.

Some examples of the use of Pu(III) for solubility measurements in nitrogen – hydrogen/carbonate mixture will be given.

Plutonium(VI) is used to study the reduction in a repository situation with the Swedish design (50 bar overpressure H₂, fuel matrix). Experiments using specially designed pressure vessels (50 bar H₂) containing solid UO₂ or TiO₂ will be presented.